

# **Growth of Laser-Induced Damage During Repetitive Illumination of HfO<sub>2</sub>-SiO<sub>2</sub> Multilayer Mirror and Polarizer Coatings**

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# Growth of laser-induced damage during repetitive illumination of $\text{HfO}_2$ - $\text{SiO}_2$ multilayer mirror and polarizer coatings

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## ABSTRACT

As designers want to increase the peak fluence of high power laser, it becomes necessary to tolerate some damage on mirrors and polarizers. To quantify how the different types of damage morphologies initiate and grow during repetitive illumination, hafnia-silica multilayer mirror and polarizer coatings were laser damage tested. The coatings were prepared by e-beam evaporation and irradiated with a 3-ns-pulse at 1064 nm.

The morphology of laser-induced damage was recorded after each shot to determine the types of damage that cause massive unstable failure and lower the optic's functional damage threshold. The results of the tests were summarized on damage stability maps plotting the average damage size as a function of the number of shots for fluences ranging from 10 to 40 J/cm<sup>2</sup>. The maps indicate that the commonly observed damage morphologies (i.e. pits, flat bottom pits, scalds and outer layer delamination) have distinct growth behaviors and influence the value of the functional damage threshold differently. While pits are stable up to fluences as high as 40 J/cm<sup>2</sup>, flat bottom pits can grow during repetitive illumination above a critical fluence of about 35 J/cm<sup>2</sup>. Scalds are formed in the first shot and never grow at fluences below 40 J/cm<sup>2</sup>. Finally, delaminates are highly unstable and have the potential for damaging the coating catastrophically above 15 J/cm<sup>2</sup>.

The results show that the delaminate damage morphology should be prevented. This knowledge has allowed coatings development efforts to focus on eliminating the origin of such damage morphology.

**Keywords:** Laser-induced damage, damage growth, damage morphology, hafnia-silica mirrors and polarizers, 1064 nm.

## 1. INTRODUCTION

The development of high power lasers such as the National Ignition Facility (NIF) at Lawrence Livermore National Laboratory (LLNL) and the Laser Mégajoules (LMJ) in France continues to challenge engineers to produce more damage resistant optical components. In the past four decades, considerable research efforts have been devoted to increasing the damage threshold of multilayer mirrors and polarizers.<sup>1-7</sup> These efforts have led to substantial improvements of the quality of such optical components by decreasing the numbers of defects in the coatings,<sup>8</sup> improving the material properties of the films (e.g. reducing the optical absorption),<sup>9-23</sup> optimizing and controlling the thin film design and spectral performance.<sup>1,24,25</sup> However, as designers are pushing the peak operating fluence of the laser to levels where defects can easily induce damage, it has become important to acknowledge that the production of large-area defect-free optics is not possible with the current technology. On the other hand, it has been observed on existing systems such as the Nova laser at LLNL and the Omega laser at the University of Rochester that damaged optics can often still operate without critical loss of performance.

Since NIF mirrors and polarizers are designed to operate at peak fluences that are higher than those of existing facilities, it is critical to quantify the effect of damage in mirrors and

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polarizers so as to experimentally determine what damage is tolerable on the laser beam line. The parameter used to quantify the maximum fluence at which an optic will not damage catastrophically, will not present damage that exceeds a critical effective size and that will not produce critical loss of reflectivity is called the functional damage threshold.<sup>26</sup> To address this issue, two series of experiments were initiated. First, the effects of damage on the characteristics of the beam (e.g. phase shifts and loss of reflectivity) were measured in order to determine the effective size of the damage.<sup>27</sup> Then, the growth of damage was characterized as a function of fluence under repetitive illumination; this study is reported in this article.

First, we will briefly discuss the four damage morphologies commonly found in mirrors and polarizers (pits, flat bottom pits, scalds and delaminates).<sup>28,29</sup> The reader can find more detailed morphological information about the different types of damage in ref. 27. The damage size as a function of number of shots and for variable fluence will then be plotted. Finally, the results will be discussed to provide general rules on the types of damage that must be avoided. After accepting that some damage is acceptable, we focused development efforts on eliminating the types of damage morphologies that lead to catastrophic failure of the optics.<sup>25</sup>

## 2. EXPERIMENTAL PROCEDURE

### 2.1 Mirror and polarizer coatings preparation

Both mirrors and polarizers were prepared by e-beam deposition of alternating layers of hafnia and silica onto 50.5 mm diameter BK7 Zygo polished substrates. The multilayers were coated at Optical Coatings Laboratory, Inc. (OCLI), Spectra Physics (S-P) and LLNL. Different sources and methods were used to deposit the film materials (e.g. hafnium oxide or metal hafnium in an oxygen partial pressure to deposit hafnia). Different sets of mirrors and polarizers were designed to operate at 45° or 56° beam angle of incidence. The coating thickness was monitored in situ by an optical monitor, switching materials by shuttering when the thickness conditions were met. The mirrors were overcoated with a half-wave silica overlayer. The polarizers were also overcoated with a very thin silica layer.

### 2.2 Laser testing conditions

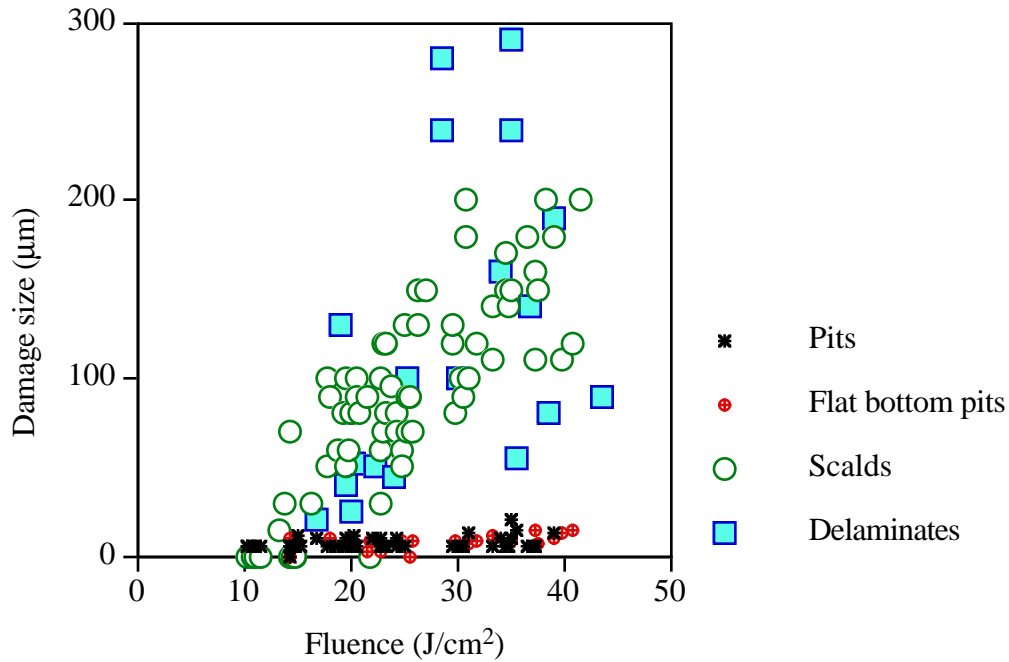
The laser damage tests were carried out using a 3-ns pulse from a 1064 nm Nd:YAG laser. The laser was focused to provide a far field circular Gaussian beam with a diameter of 1.1 mm at  $1/e^2$  of the maximum intensity. The beam profile was recorded for each shot (except for S-on-1 tests) and the peak fluence was computed. Each site was irradiated under 1-on-1 (one laser pulse), N-on-1 (N successive shots) and S-on-1 (600 shots at a 10 Hz repetition rate) conditions. The tests were conducted in s- or p-polarization at use angle. Two sets of mirrors were designed to reflect at 45° or 56°. The polarizers were designed to operate at 56°. A total number of 6 mirrors and 2 polarizers were tested. An average of 30 sites were irradiated on each sample. The sites were spaced 5 mm from one another to avoid conditioning effects from adjacent site illumination and minimize potential cross-contamination. The mirrors and polarizers were tested at fluences ranging from 10 to 45 J/cm<sup>2</sup>. The samples were examined before and after irradiation by Nomarski and back light microscopy. Any damage larger than 2  $\mu$ m was detected. Further post damage characterization was conducted using scanning electron microscopy (SEM).

## 3. RESULTS

An earlier article<sup>27</sup> reported the damage behavior of hafnia-silica mirrors and polarizers after 1-on-1 tests. Figure 1 shows the average damage size for the different types of morphologies as a function of fluence. This figure summarizes the damage results for mirrors and polarizers from various origins tested in s- and p-polarization. It indicates that

the general dependence of the size of each type of morphology on fluence is quite consistent. The number of damage sites can vary with process, optic type or vendor, but the general behavior of a given damage morphology is quite reproducible.

The damage growth study showed that the growth behavior of a given damage morphology is also typical. This section will therefore present the general behavior of each type of damage morphology (rather than focusing on one type of sample) and will present plots of damage size as a function of number of shots and fluence with Nomarski optical micrographs representative of typical damage phenomena. Indeed, quantitative differences (e.g. value of the functional damage threshold) are expected to vary from sample to sample or for optical components with different optical designs, coating process or tested in different conditions.

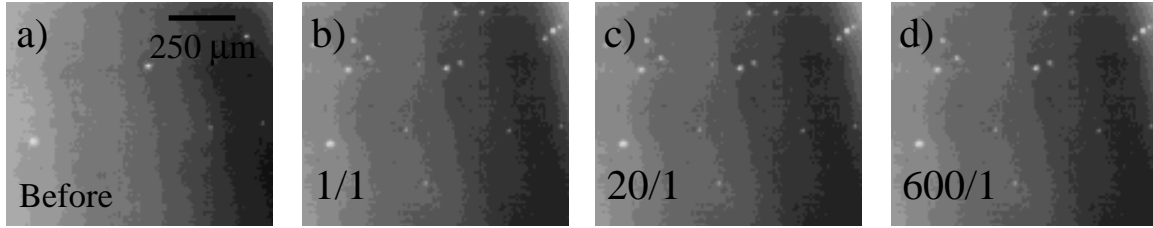


**Fig. 1:** Damage size of the pit, flat bottom pit, scald, and delaminate morphologies as a function of fluence after a single shot (from ref. 27). The measurements for the mirrors and polarizers of various origins (i.e. OCLI, S-P, LLNL) tested at 45° and 56° are plotted on this graph.

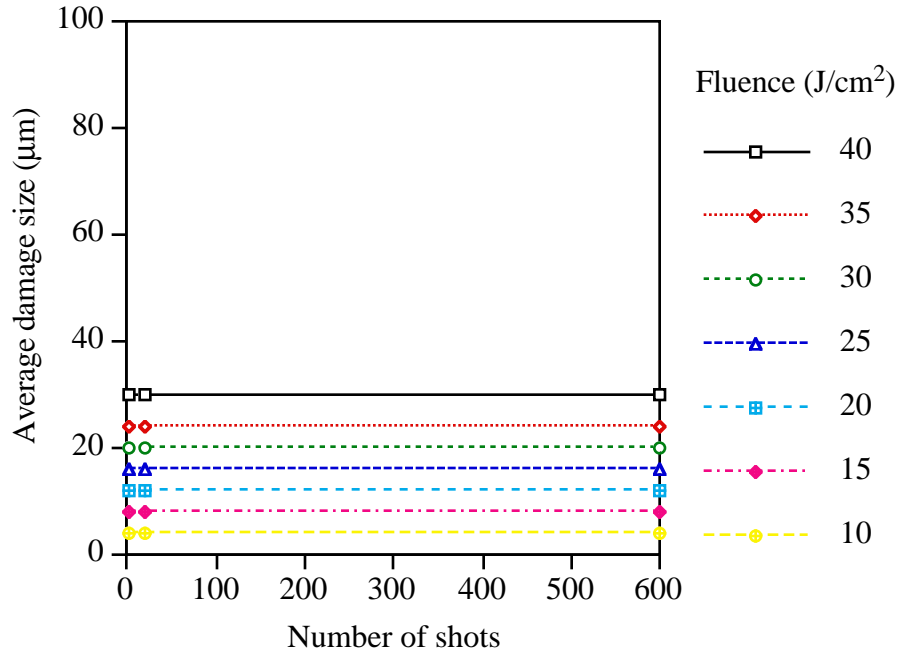
### 3.1 Pits

Pits formation usually occurs during the initial shot by ejection of nodular defects.<sup>30,31</sup> The damage tests on the reflective optics showed that the pit morphology does not tend to grow during repetitive illumination (see Figs. 2 and 3). The pit size does not usually exceed 40 μm. Although it is difficult to quantify exactly the increase of pit size with fluence because of the lack of resolution with the optical microscope, pits were generally found to be smaller when the samples are initially irradiated at fluences below 15 J/cm². This agrees with previous experimental studies reporting that initial low fluence illumination or laser conditioning can improve the damage threshold.<sup>32,33</sup> This study also suggests that from a functional damage improvement standpoint, multiple step conditioning could be replaced by a single step conditioning process.

The results shown in this article *only apply to reflectors*; other tests on polarizers in p-polarization have shown that large (>60 μm) and deep pits can form at fluences near 30 J/cm². These large pits can be unstable during repetitive illumination.



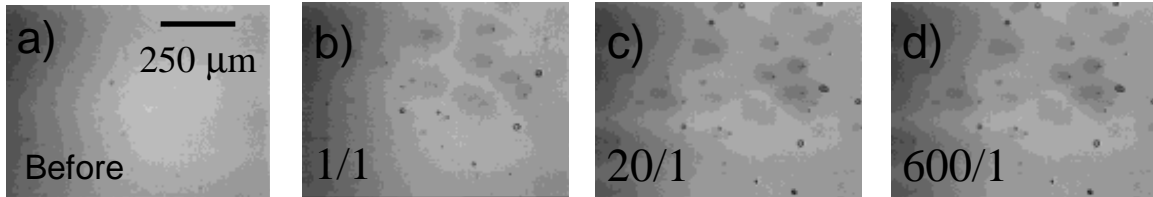
**Fig. 2:** Back light optical micrographs of pits a) before, after b) 1, c) 20 and d) 600 shots at 1064 nm on a mirror at  $40 \text{ J/cm}^2$  at  $45^\circ$ , in p-polarization.



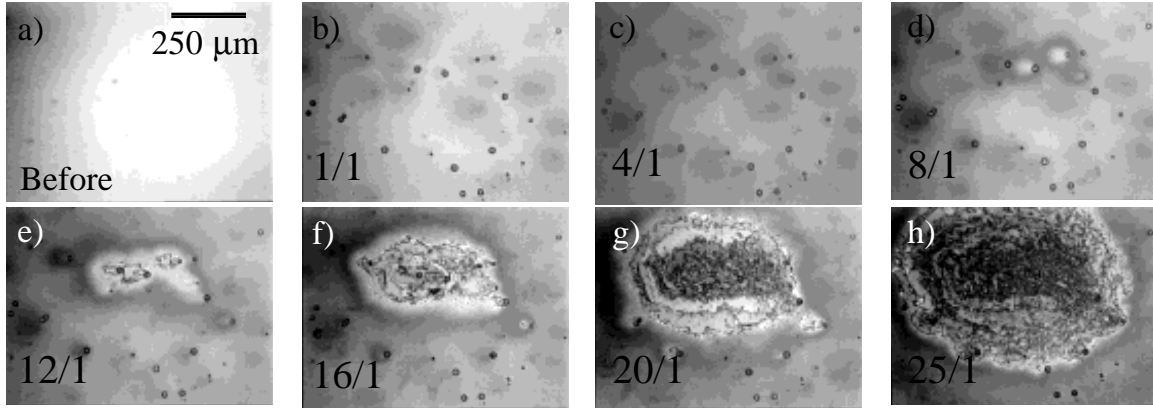
**Fig. 3:** Average pit size as a function of number of shots at 1064 nm in  $45^\circ$  mirrors tested in p-polarization at fluences ranging from 10 to  $40 \text{ J/cm}^2$ .

### 3.2 Flat bottom pits

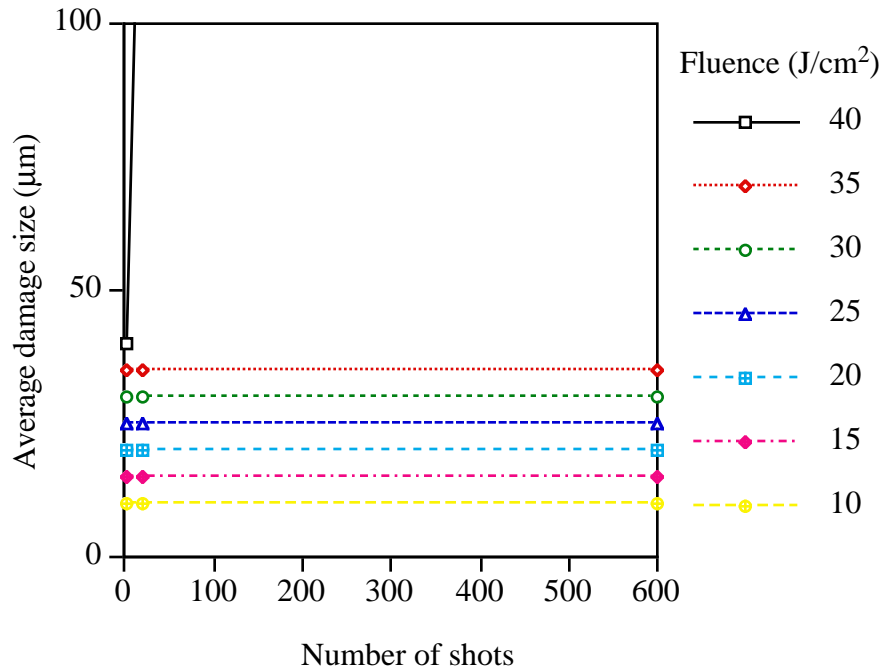
Flat bottom pits were occasionally found in some of the optics studied. SEM characterization of this type of damage showed that the top layer can fail at the hafnia-silica interface.<sup>34</sup> Figures 4 and 5 show the typical damage growth behaviors of flat bottom pits at 31 and  $44 \text{ J/cm}^2$  in a high reflector coating. The damage size after one shot increases with fluence. Such damage does not grow at low fluences but can become unstable above a critical fluence (see Fig. 6). It is possible that damage growth depends on the initial size of the pit; this effect was not investigated in this study.



**Fig. 4:** Nomarski optical micrographs of an irradiated site a) before, and after b) 1, c) 20 and d) 600 shots at 1064 nm on a mirror. The flat bottom pits shown here were formed at  $31 \text{ J/cm}^2$  at  $45^\circ$ , in p-polarization and did not grow.



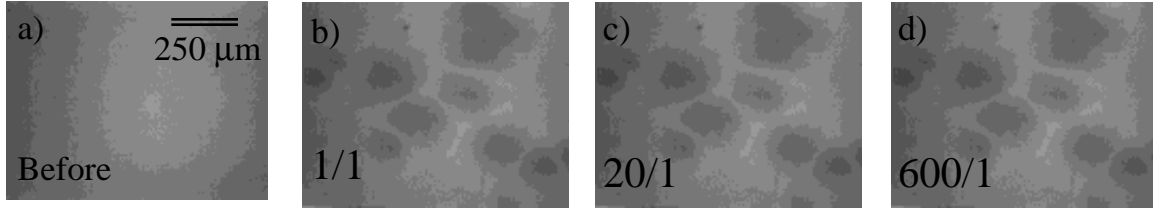
**Fig. 5:** Nomarski optical micrographs of an irradiated site a) before, and after b) 1, c) 4, d) 8, e) 12, f) 16, g) 20 and h) 25 shots at 1064 nm on the same mirror as Fig. 4. The flat bottom pits shown here were formed at 44 J/cm<sup>2</sup> at 45°, in p-polarization and grew catastrophically.



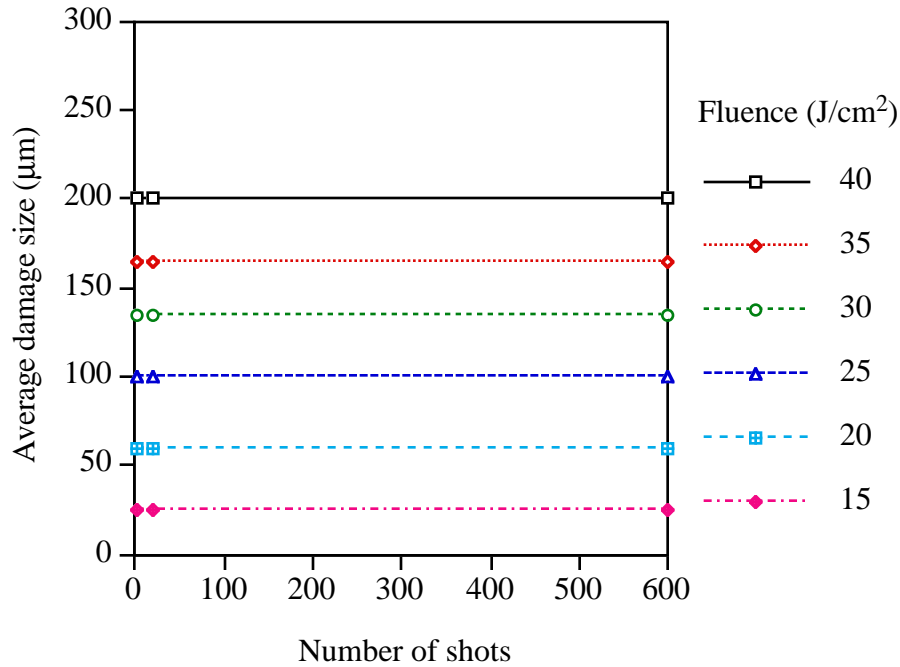
**Fig. 6:** Average flat bottom pit size as a function of number of shots at 1064 nm in 45° mirrors tested in p-polarization at fluences ranging from 10 to 40 J/cm<sup>2</sup>.

### 3.3 Scalds

Scalds were typically found at fluences above 13 J/cm<sup>2</sup>. Their size increased almost linearly with fluence. It is believed that scalds result from plasmas that ignite at nodular defects or contamination particles and scorch the silica surface. The earlier study of the 1-on-1 damage morphologies<sup>27</sup> showed that such damage can only produce very small phase modulations and therefore do not affect the functional damage threshold of the high reflectors. The scalds are formed on the first shot and do not grow further even at high fluences (see Figs. 7 and 8). Their size increases with pulse length and fluence.



**Fig. 7:** Nomarski optical micrographs of an irradiated site a) before, and after b) 1, c) 20 and d) 600 shots at 1064 nm on a mirror. The scalds shown here formed on the first shot at 41 J/cm<sup>2</sup> at 45°, in p-polarization and did not grow.



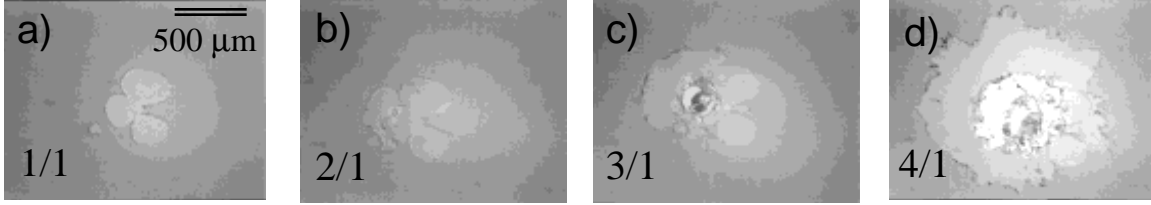
**Fig. 8:** Average scald size as a function of number of shots at 1064 nm in 45° mirrors tested in p-polarization at fluences ranging from 15 to 40 J/cm<sup>2</sup>.

### 3.4 Delaminates

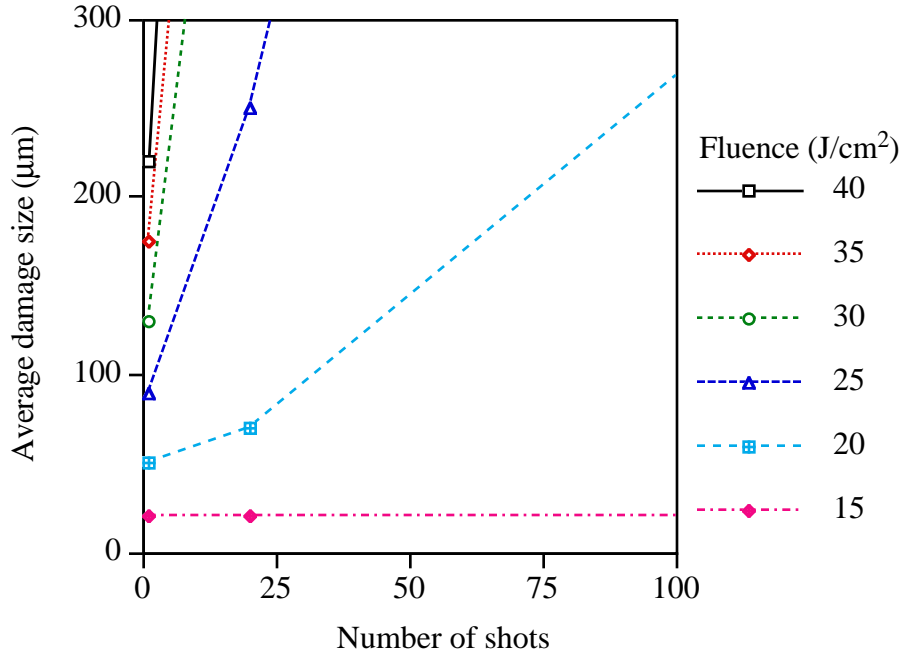
Delaminates (i.e. delamination of the outer layer) have characteristics that are similar to scalds; they seem to occur as a result of plasma ignition and the size vs. fluence relationship is very similar to that of scalds. Delaminates can very quickly grow to catastrophic proportions (see Figs. 9 and 10). They represent the type of damage that most limits the functional threshold of the high reflectors, and particularly polarizers.<sup>25,27</sup>

The recent development effort of NIF polarizers focused on eliminating the cause of delamination. Other research conducted in parallel with this study also showed that the electric field distribution in the multilayer stack (which can vary with thin film design or beam incidence angle) can strongly influence the damage morphology. Such effects are reported elsewhere in these proceedings.<sup>25,35</sup>





**Fig. 9:** Nomarski optical micrographs of delaminates after a) 1, b) 2, c) 3, and d) 4 shots at 1064 nm on a polarizer. The damage grew very rapidly to catastrophic proportions only after 4 shots at  $46 \text{ J/cm}^2$  at  $54^\circ$ , in s-polarization.



**Fig. 10:** Average delaminate size as a function of number of shots at 1064 nm in  $56^\circ$  polarizers tested in s-polarization at fluences ranging from 15 to  $40 \text{ J/cm}^2$ .

#### 4. CONCLUSION

As designers want to increase the peak fluence of high power laser such as NIF and LMJ, it becomes necessary to tolerate some damage on mirrors and polarizers. To quantify the survivability of damaged optical components, the growth of the different types of damage morphologies was studied during repetitive illumination. Hafnia-silica multilayer mirror and polarizer coatings were irradiated with a 3-ns pulse at 1064 nm. The morphology of laser-induced damage was recorded by optical microscopy after each shot to build damage stability maps plotting the average damage size as a function of the number of shots for fluences ranging from 10 to  $40 \text{ J/cm}^2$ .

The four damage morphologies commonly found were investigated. The results show that each type of damage morphology has a particular growth behavior. While pits seem to be stable up to  $40 \text{ J/cm}^2$ , flat bottom pits (which are generally larger) can induce catastrophic failure above a critical fluence (typically  $35 \text{ J/cm}^2$ ). Scalds and delaminates were found to be associated with plasma formation. While scalds were very stable, delaminates grew very quickly to catastrophic proportions above  $20 \text{ J/cm}^2$  leading to rather low values of the functional damage threshold. The occurrence of the delaminate

morphology was found to be sensitive to the thickness of the silica overlayer. Their occurrence was prevented by increasing the thickness of the overlayer.

By characterizing the damage morphology of a sample, it is now possible to predict the fluence range at which a given mirror or polarizer can operate without critical failure. In general, the formation of delaminates translates into destruction of the coating at relatively low fluences while other types of damage morphologies do not limit the laser operation at fluences up to about 35 J/cm<sup>2</sup>.

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